FORM PTO-1390 (REV 1-98) U.S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER: 005317-20009

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NUMBER (if known, see 37 C.F.R. 1.5): 09/29[illegible]83 NOT ASSIGNED

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/JP98/03935	September 2, 1998	September 2, 1997

TITLE OF INVENTION: ORGANIC EL ELEMENT MANUFACTURING PROCESS AND ORGANIC EL ELEMENT

APPLICANT(S) FOR DO/EO/US: SEKI, Shunichi; KIGUCHI, Hiroshi both of Nagano-Ken, Japan

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☒ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:

U.S. APPLICATION NO. (if known, see 37 C.F.R. 1.5)	INTERNATIONAL APPLICATION NUMBER	ATTORNEY'S DOCKET NUMBER
09/457483 NOT ASSIGNED	PCT/JP98/03935	005317-20009

17. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 C.F.R. 1.492(a)(1)-(5)):

Neither international preliminary examination fee (37 C.F.R. 1.482) nor international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO **$970**

International preliminary examination fee (37 C.F.R. 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO **$840**

International preliminary examination fee (37 C.F.R. 1.482) not paid to USPTO but international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO **$760**

International preliminary examination fee (37 C.F.R. 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$670**

International preliminary examination fee (37 C.F.R. 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) **$96**

CALCULATIONS PTO USE ONLY	
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 970
Surcharge of **$130** for furnishing the oath or declaration later than ☒ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. 1.492(e)).	$ 130

CLAIMS	NUMBER FILED	NUMBER ALLOWED	NUMBER EXTRA	RATE	
Total claims	68	20	48	$ 18	864
Independent claims	8	3	3	$ 78	234
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+	**$260**	$ 260
TOTAL OF ABOVE CALCULATIONS =					2458
Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also by filed (Note 37 C.F.R. 1.9, 1.27, 1.28).					
SUBTOTAL =					$2,458
Processing fee of **$0** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 C.F.R. 1.492(f)).				+	$ 0
TOTAL NATIONAL FEE =					2458
Fee for recording the enclosed assignment (37 C.F.R. 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31). **$40** per property				+	$ 0
TOTAL FEES ENCLOSED =					2458
$					**Amt. Refunded** $ **Amt. charged**

a. ☒ A check in the amounts of 2458 to cover the above fees are enclosed.

b. ☐ Please charge my Deposit Account No. 12-1820 in the amount of 0 to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 12-1820. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

LOEB & LOEB LLP
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Form PTO-1390 (REV 1-98)

SIGNATURE

William H. Wright
REG.NO. 36,312 EL 340 695 936 US
April 30, 1999

Transmittal Letter to the United States Designated Office (DO/US)—Entry Into National Stage under 35 U.S.C. 371—PTO 1390 **[13-7]**

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PATENT
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:	Art Unit: NOT ASSIGNED
SEKI, Shunichi; KIGUCHI, Hiroshi	Examiner: NOT ASSIGNED
Serial No: NOT ASSIGNED	
Filed: April 30, 1999	
For: ORGANIC EL ELEMENT MANUFACTURING PROCESS AND ORGANIC EL ELEMENT	

PRELIMINARY AMENDMENT

Box PCT
Assistant Commissioner for Patents
Washington, D.C. 20231

Dear Sir:

Prior to the first office Action in the present application, please enter and consider the following amendments and remarks:

IN THE CLAIMS:

Claim 9, line 2, replace "8", with --7--.

Claim 14, line 3, replace "13", with --7--.

Claim 16, lines 2 and 3, replace "any of Claims 1 through 15", with --Claim 1--.

Claim 17, line 12, replace "any of Claims 1 through 16", with --Claim 1--.

REMARKS

Entry of this Amendment is respectfully requested to make claims more consistent U.S. Practice.

If for any reason the Examiner finds the application other than in condition for allowance, the Examiner is requested to call the undersigned attorney at the Los Angeles, California telephone number 310-282-2000 to discuss the steps necessary for placing the application in condition for allowance.

Respectfully submitted,

LOEB & LOEB LLP

Date: April 30, 1999

By: ____________________
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SPECIFICATION

ORGANIC EL ELEMENT MANUFACTURING PROCESS AND ORGANIC EL ELEMENT

TECHNICAL FIELD

The present invention relates to a manufacturing process for an organic EL (electroluminscence) element, an electrically light-emitting element that may be used in displays, display light sources, and the like. In particular, it relates to a composition for use as a hole injecting and transporting layer suitable for ink jet patterning.

BACKGROUND ART

When electric charge is applied to a solid thin film containing a fluorescent organic molecule and sandwiched between electrodes, holes (holes) are injected from the anode and electrons from the cathode, these carriers moving through the thin film due to the applied electric field, and recombining. An element in which the energy released during this recombination is consumed in the formation of a singlet excitation state of the fluorescent molecule (molecule exciton), and the

fluorescence emitted with relaxation of the singlet exciton to ground state is utilized is an organic EL element.

Apropos, with a single-layer type structure element comprising a light-emitting layer only, light emission efficiency is low and there are problems with durability, and therefore a two-layer type structure element provided with a hole injecting and transporting layer with good contact situated between the anode and the light-emitting layer has been proposed. Through the use of a stacked structure to control carrier injection/transport balance and carrier recombination location, EL light-emitting element light emission efficiency and durability can be improved. Further, according to a stacked structure, the functions of light emission and injection/transport can be carried out by separate materials, which provides the advantage of making possible optimal design of materials and elements.

In the past, as hole injecting and transporting layer compounds for two-layer stacked type organic EL elements, there have been proposed in the past a porphyrinic compound (US Patents 4,356,429 and 4,720,432), aniline, pyridine, and low-molecular weight derivatives thereof (Unexamined Patent Application (Kokai) 3-34382), hole injecting and transporting layers using carbon layers (Unexamined Patent Application (Kokai) 8-31573),

etc. The formation of hole injecting and transporting layers using these low-molecular weight materials is usually accomplished by film formation methods involving vacuum deposition or sputtering. Polyaniline (Nature, 357, 477 (1992), etc. are known as low-molecular weight materials, film formation being carried out by a wet process such as spin coating, etc.

However, film formation methods involving vacuum deposition or sputtering are batch processes having extended time periods, and thus mass production efficiency is poor. In the case of low-molecular weight materials, there are the problems of susceptibility to crystallization after film formation and lower element reliability. On the other hand, in the case of high-molecular weight materials, there are the advantages of a high degree of freedom in molecular design, and ease of optimization of materials owing to a wet process; however, film formation methods such as spin coating, etc., have a big problem in that virtually all of the material is wasted.

In cases where fine patterning of materials is required, as in full-color displays, etc., there are the fundamental problems that high-precision patterning is difficult in deposition processes, while the materials lack durability against patterning processes involving photolithography. The same problems exist in the case of

high-molecular weight materials as well. The materials used as hole injecting and transporting layers or buffer layers are conductive, and thus if complete patterning cannot be realized, this can result in leakage current between adjacent pixels located on a given substrate.

DISCLOSURE OF THE INVENTION

Accordingly, it is an object of the present invention to provide a composition for hole injecting and transporting layers that allows optimization of material and element design to be carried out, and that allows high-precision patterning film formation to be carried out easily, in a short time, and at low cost. It is a further object to provide a manufacturing process for a composition for hole injecting and transporting layers. It is yet a further object to provide a manufacturing process for an organic EL element using this composition. It is yet a further object to provide an organic EL element manufactured by means of this manufacturing process and having excellent light emission characteristics.

The composition for a hole injecting and transporting layer which pertains to the present invention is a composition for use in patterning formation of a hole injecting and transporting layer of

an organic EL element using an ink-jet type recording head, contains a polar solvent for dispersing or dissolving a conductive compound for forming the hole injecting and transporting layer and a lubricant for facilitating ink-jet patterning, and has the following physical properties (contact angle, viscosity, surface tension).

(1) Contact angle

In preferred practice, the contact angle of the composition for a hole injecting and transporting layer and the material making up the nozzle face of the ink-jet type recording head will be established within the range 30 °-170 °. Establishment within the range 35 °-65 ° is especially preferred.

By means of the composition for a hole injecting and transporting layer having a contact angle of this range, bending of flight during discharge can be reduced, making possible precise discharge control. Where the contact angle is smaller than 30 °, wetting of the composition for a hole injecting and transporting layer at the nozzle face increases, so that when the composition for a hole injecting and transporting layer is discharged, the composition for a hole injecting and transporting layer sometimes adheres asymmetrically about the perimeter of the nozzle hole. In this case, mutual attraction between the composition for a hole injecting and transporting

layer adhering to the nozzle and the composition for a hole injecting and transporting layer which is being discharged acts in such a way that the composition for a hole injecting and transporting layer is discharged through uneven force, so that bending of flight occurs and the target location cannot be impacted. Further, the frequency of bending of flight increases. On the other hand, where the contact angle exceeds 170 °, interaction between the composition for a hole injecting and transporting layer and the nozzle hole is at a minimum, and the shape of the meniscus at the nozzle distal end is not stable, with the result that the composition for a hole injecting and transporting layer discharge amount and discharge timing are difficult to control.

Bending of flight refers to misplacement of the impact location of the composition for a hole injecting and transporting layer by 30 μm or more away from the target location when the composition for a hole injecting and transporting layer is discharged from the nozzle hole. Frequency of bending of flight refers to the time elapsed until bending of flight occurs during continuous discharge at an ink-jet type recording head piezoelectric thin film element oscillation frequency of 14.4 kHz, for example.

(2) Viscosity

In preferred practice, the viscosity of the composition for a hole injecting and transporting layer will be in the range 1 cps-20 cps. Establishment within the range 2 cps-4 cps is especially preferred.

Where the viscosity of the composition for a hole injecting and transporting layer is below 1 cps, the meniscus of the composition for a hole injecting and transporting layer at the nozzle hole will not be stable, making discharge control difficult. On the other hand, where the viscosity exceeds 20 cps, it will not be possible to smoothly discharge the composition for a hole injecting and transporting layer from the nozzle hole, making ink discharge difficult unless the specifications of the ink-jet type recording head are modified, such as by making the nozzle hole bigger, etc. Where viscosity is high, the solid ingredients in the composition for a hole injecting and transporting layer will tend to precipitate, increasing the frequency of nozzle hole clogging.

(3) Surface tension

In preferred practice, the surface tension of the composition for a hole injecting and transporting layer will be established in the range 20 dyne/cm-70 dyne/cm. Establishment within the range 25 dyne/cm-40 dyne/cm is especially preferred.

By establishing the surface tension within this range, bending of flight is reduced analogously to the contact angle described earlier, and the frequency of bending of flight can be reduced. Where surface tension is greater than 70 dyne/cm, the shape of the meniscus at the nozzle distal end is not stable, with the result that the composition for a hole injecting and transporting layer discharge amount and discharge timing are difficult to control. On the other hand, where surface tension is lower than 20 dyne/cm, wetting of the composition for a hole injecting and transporting layer with respect to the material of the nozzle face increases, so that, as with the contact angle described earlier, bending of flight occurs and the frequency of bending of flight becomes high.

This bending of flight occurs mainly when there is uneven wetting of the nozzle hole, when there is clogging due to adhesion of solid matter in the composition for a hole injecting and transporting layer, etc., and can be solved through cleaning of the ink-jet type recording head (hereinafter termed "flushing"). Flushing usually involves working the ink-jet type recording head mechanism so as to prevent clogging and bending of flight, the design being such that a prescribed amount of the composition for a hole injecting and transporting layer is forcedly discharged when discharge of the composition

for a hole injecting and transporting layer has not been carried out for a given time (hereinafter termed "flushing time"). This flushing time refers to the time required for a nozzle to dry and give rise to bending of flight when not discharging the composition for a hole injecting and transporting layer, and is indicative of the properties of the composition for a hole injecting and transporting layer. The longer the flushing time, the more appropriate [the composition is] for ink-jet printing techniques, thus allowing the composition for a hole injecting and transporting layer to be discharged in a stable manner for extended periods.

Accordingly, by endowing the composition for a hole injecting and transporting layer with the physical properties mentioned earlier, it is possible to produce a longer flushing time and to maintain a more fresh state at the interface of the composition for a hole injecting and transporting layer with the air. It is additionally possible to produce uniform dot density of the composition for a hole injecting and transporting layer being discharged, thereby making it possible to prevent the occurrence of irregularities, etc., of the composition for a hole injecting and transporting layer. Further, since the linearity of flight is excellent, control of the ink-jet type recording head is facilitated,

and a simple constitution may be adopted for the manufacturing equipment.

In preferred practice, a mixed solution of water and a lower alcohol (e.g., methanol, ethanol, etc.) will be used as the polar solvent. A mixed solution of water and a Cellosolve solvent (e.g., ethoxy ethanol, etc.) can also be used. In preferred practice, the lubricant will be glycerin. The manufacturing process for a composition for a hole injecting and transporting layer which pertains to the present invention may entail preparation via a sonication process and a filtration process.

The manufacturing process for an organic EL element which pertains to the present invention is a manufacturing process for an organic EL element having a stacked structure of a hole injecting and transporting layer and a light-emitting layer formed within a partitioning member divided into individual pixel areas, comprising a step wherein a partitioning member provided with openings corresponding to pixel areas is formed on a substrate; a step wherein an ink-jet type recording head is used to fill the aforementioned openings with the composition for a hole injecting and transporting layer which pertains to the present invention; and a step wherein the composition for a hole injecting and transporting layer that has filled the openings is subjected to a drying process to form a hole injecting

and transporting layer. Through this method, hole injecting and transporting layer film thickness, dot count, and other parameters may be controlled in a desired manner, allowing the size and pattern of the organic EL element to be established in a desired manner as well.

The organic EL element which pertains to the present invention is manufactured through the foregoing process, and the film thickness of the hole injecting and transporting layer is 0.1 μm or less. The film resistance of the hole injecting and transporting layer is in the range 0.5×10^9 Ω/m² to 5×10^9 Ω/m². By establishing the film thickness and the film resistance of the hole injecting and transporting layer within the preceding ranges, the light-emission characteristics of the organic EL element can be improved.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a sectional view of manufacturing steps for an organic EL element.

BEST MODE FOR CARRYING OUT THE INVENTION

Embodiment of Invention 1.

A composition for a hole injecting and transporting layer, organic EL element manufacturing process, and organic EL element pertaining to an aspect of the present embodiment is described below.

The composition for a hole injecting and transporting layer principally contains a conductive compound for forming the hole injecting and transporting layer, a dispersant solvent, and a lubricant, and is used for pattern film formation through an ink-jet system. In preferred practice, the conductive compound for forming the hole injecting and transporting layer is a compound having lower ionization potential than the anode. For example, where ITO is used for the anode, examples of low-molecular weight materials would include copper phthalocyanine and other porphyrinic compounds.

Other additives, as well as film stabilizing materials, may be added as well; for example, viscosity modifiers, antioxidants, pH modifiers, preservatives, resin emulsions, leveling agents, etc., may be used.

(Example)

Using copper phthalocyanine as the conductive compound (hole injecting and transporting layer component), the physical characteristics of compositions for a hole injecting and transporting layer [produced therefrom] were studied. Composition 1 through

Composition 10, given in Table 1 through Table 10, were prepared as specimens.

Table 1

Composition 1

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	70
Lubricant	glycerin	5

Table 2

Composition 2

	Ingredient	Content (wt%)
Hole injecting and transporting layer component	copper phthalocyanine (10 wt%)(styrene acrylic resin dispersion)	25
Polar solvent	water	65
	methanol	5
Lubricant	glycerin	5

Table 3

Composition 3

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	65
	ethoxy ethanol	5
Lubricant	glycerin	5

Table 4

Composition 4

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	methanol	70
Lubricant	glycerin	5

Table 5

Composition 5

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	N, N-methyl formamide	70
Lubricant	glycerin	5

Table 6

Composition 6

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	70
Lubricant	--	0

Table 7

Composition 7

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	70
	methanol	5
Lubricant	--	0

Table 8

Composition 8

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	70
	ethoxy ethanol	5
Lubricant	--	0

Table 9

Composition 9

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	65
	butoxy ethanol	5
Lubricant	glycerin	5

Table 10

Composition 10

	Ingredient	Content (wt%)
Hole injecting and transporting layer Component	copper phthalocyanine (10 wt%) (styrene acrylic resin dispersion)	25
Polar solvent	water	65
	isopropyl alcohol	5
Lubricant	glycerin	5

(Discharge Evaluation)

Composition 1 through Composition 8, given in Table 1 through Table 8, were measured for contact angle with respect to the nozzle face constituent material constituting the ink-jet type recording head, viscosity, and surface tension, and the discharge properties thereof were evaluated. The discharge evaluation was conducted using an ink-jet printing device (Epson MJ-500C).

Viscosity [is expressed as] measured values at 20°C. These results are given in Table 11.

Table 11

Composition	Contact angle [°]	Viscosity [cps]	Surface tension [dyne/cm]	Discharge properties
1	135	3.8	62.8	O
2	91	3.6	40.8	O
3	62	3.1	39.8	◎
4	22	0.8	23.1	X
5	175	0.9	81.0	X
6	118	1.1	71.0	X
7	28	0.8	68.8	X
8	27	0.9	69.2	X

From these results, it will be apparent that a contact angle of 30° to 170°, and especially 35° to 65°, is preferred. Further, viscosity of 1 cps to 20 cps, and especially 2 cps to 4 cps is preferred, and surface tension of 20 dyne/cm to 70 dyne/cm, and especially the range 25 dyne/cm to 40 dyne/cm, is preferred.

Composition 1 through Composition 3, which contain glycerin as a lubricant, when compared to Composition 6 through Composition 8, which do not contain a lubricant, may be seen to be superior in terms of discharge properties. Accordingly, in preferred practice, the ink composition will contain a lubricant. The inclusion of a lubricant can effectively prevent drying and solidification of the ink composition in the nozzle mouth. Examples of the lubricant are glycerin, diethylene glycol,

and other polyhydric alcohols; glycerin is especially preferred.

(Manufacturing process for composition for a hole injecting and transporting layer)

Composition 1 through Composition 3, Composition 9, and Composition 10 given in Table 1 through Table 3, Table 9, and Table 10, respectively, were manufactured, and the particle size distribution of the hole injecting and transporting layer-forming compound (copper phthalocyanine) was measured before and after a sonication. Using the preceding compositions for a hole injecting and transporting layer having been passed through a filtration step subsequent to the sonication , the film forming properties of hole injecting and transporting layers formed through ink-jet system patterning were evaluated.

These results are given in Table 12. Sonication results are expressed as the proportion of the particle size distribution of 1 μm or smaller.

Particle size in the styrene acrylic resin dispersion was 1 μm or above.

Table 12

Composition	1 μm particle size proportion (%)		Film forming properties
	before sonication	after sonication	
1	4.8	46.8	○
2	2.8	31.4	○
3	4.2	43.5	◎
9	2.5	18.5	X
10	3.9	18.2	X

From the preceding results, it will be apparent that 4-hour sonication of the dispersion can increase dispersion. Further, by additionally filtering the sonicated dispersion, it is possible to achieve a more homogeneous hole injecting and transporting layer film. As the dispersant polar solvent for the conductive compound, water or a mixed solvent of water and methanol or ethoxy ethanol is preferred (Composition 1 through Composition 3); it will be apparent that, where these solvents are used, film forming properties are good.

(Organic EL element manufacturing steps)

Using Composition 1 through Composition 3 given in Table 1 through Table 3, patterning film formation of hole injecting and transporting layers by an ink-jet

system was carried out by the following procedure to manufacture organic EL elements.

Anode formation step (Fig. 1A)

The present step is a step for forming an anode 101 on a glass substrate 102. In preferred practice, glass substrate 102 will be one that resists attack by chemicals such as acids, alkalis, etc., and that is mass-producible. An ITO transparent electrode was formed to a thickness of 0.1 μm on substrate 102 and patterned at 100 μm pitch.

Partitioning member formation step (Fig. 1B)

The present step is a step for forming a partitioning member 103 on glass substrate 102. Specifically, non-photosensitive polyimide (partitioning member) buried between anodes (ITO electrodes) and also serving as an ink drip prevention wall (bank) was formed by photolithography. The non-photosensitive polyimide was 20 μm wide and 2.0 μm thick.

Composition for a hole injecting and transporting layer discharge step (Fig. 1C)

Hole injecting and transporting layer Compositions 1 through 3 (106 in the drawing) were discharged from the head 105 of an ink-jet printing device (Epson MJ-800C) 104 to effect patterning film formation of a hole injecting and transporting layer 107. After patterning

film formation, a hole injecting and transporting layer was formed through a drying treatment (200°C, 10 min). During discharge of the composition for a hole injecting and transporting layer, no bank-overflowing application was observed, and a high-precision hole injecting and transporting layer pattern was achieved.

Light-emitting layer composition filling step (Fig. 1D)

Next, a PPV precursor (poly(para-phenylene vinylene)) composition was manufactured as a green light-emitting layer. A light-emitting layer composition 108 was discharged by an ink-jet system to effect patterning film formation of a light-emitting layer 109. Rhodamine B-doped PPV, which exhibits red light emission, or coumarin-doped PPV, which exhibits blue light emission, could be used for the light-emitting layer 109 as well. Light-emitting layers exhibiting light emission of the three primary colors (red, green, blue) can be subjected to patterning over a hole injecting and transporting layer to produce a highly detailed full-color organic EL display.

Cathode formation step (Fig. 1E)

Finally, a cathode 100 was deposited so as to cover the light-emitting layer 109, forming an organic EL element.

(Hole injecting and transporting layer film formation evaluation)

In the organic EL element manufacturing steps described above, the number of discharges of the composition for a hole injecting and transporting layer was varied, and the hole injecting and transporting layer film thickness and sheet resistance were measured, as well as evaluating film formation. Results are given in Table 13.

Table 13

Composition	Discharge no.	Film thickness [μm]	Resistance [Ω/m²]	Film-forming properties	Luminance [cd/m²]
1	1	0.05	5.0×10^9	O	2000
	3	0.14	1.7×10^9	O	1900
	5	0.26	0.9×10^9	X	1600
2	1	0.03	0.7×10^9	◎	2000
	3	0.1	0.2×10^9	X	1500
	5	0.14	0.1×10^9	X	1500
3	1	0.02	0.5×10^9	◎	2000
	3	0.06	0.2×10^9	O	1800
	5	0.1	0.1×10^9	X	1400

From these results, it will be apparent that in the case of a low-molecular weight material, where the hole injecting and transporting layer film thickness is 0.05 μm or smaller and film resistance is from 0.5×10^9 Ω/m^2 to 5×10^9 Ω/m^2, light-emission characteristics are good.

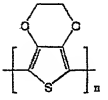
(Effect)

According to the present aspect of the embodiment, fine patterning can be realized easily, in a short time, and at low cost. Accordingly, there is no risk of leakage due to the hole injecting and transporting layer itself, a problem that could not be solved in solid film formation methods. Further, since film thickness can be readily controlled through manipulation of the discharge amount or number of discharges, optimization of thin film design thereby is possible.

Aspect of Embodiment of the Invention 2.

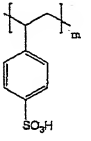
The present aspect of embodiment is one in which the conductive compound for forming the hole injecting and transporting layer is a material capable of forming a film from solution (high-molecular weight material), and is one that employs polyaniline, polysilane, or other conductive high-molecular weight material. In particular, as water can be used as the principal solvent, and as characteristics can be adjusted through the mixture ratio, a mixed material of PEDT (polyethylene dioxythiophene)

Chemical Formula 1



and PSS (polystyrene sulfonic acid)

Chemical Formula 2



is preferred.

(Example)

Compositions for a hole injecting and transporting layer prepared using a PEDT/PSS mixture aqueous solution as the high-molecular weight material (hole injecting and transporting layer component) were prepared in the five varieties (Composition 11 through Composition 15) given in Table 14 through Table 18.

Table 14

Composition 11

Composition	Material name	Content (wt%)
Hole injecting and transporting layer component	PEDT/PSS mixture aqueous dispersion(1.0 wt%)	20
Polar solvent	water	80
Lubricant	--	0

Table 15

Composition 12

Composition	Material name	Content (wt%)
Hole injecting and transporting layer component	PEDT/PSS mixture aqueous dispersion(1.0 wt%)	20
Polar solvent	N,N-dimethyl formamide	75
Lubricant	glycerin	5

Table 16

Composition 13

Composition	Material name	Content (wt%)
Hole injecting and transporting layer component	PEDT/PSS mixture aqueous dispersion(1.0 wt%)	20
Polar solvent	water	75
Lubricant	glycerin	55

Table 17

Composition 14

Composition	Material name	Content (wt%)
Hole injecting and transporting layer component	PEDT/PSS mixture aqueous dispersion(1.0 wt%)	20
Polar solvent	water	70
	methanol	5
Lubricant	glycerin	5

Table 18

Composition 15

Composition	Material name	Content (wt%)
Hole injecting and transporting layer component	PEDT/PSS mixture aqueous dispersion(1.0 wt%)	20
Polar solvent	water	65
	ethoxy ethanol	10
Lubricant	glycerin	5

Discharge evaluations were conducted using an ink-jet printing device (Epson MJ-800C). Film-forming properties evaluations were conducted in film state after a heating treatment (200°C, 10-60 min) conducted after application through discharge. Viscosity [is expressed as] measured values at 20°C.

Table 19

Composition	Contact angle[°]	Viscosi [cps]	Surface tension [dyne/cm]	Discharge properties	Film-forming properties
1	130	1.2	73.0	X	–
2	63	0.9	38.5	X	–
3	120	4.5	68.0	O	Δ
4	84	4.0	62.3	◎	O
5	64	3.5	40.0	◎	◎

From the preceding results, it will be apparent that the concentration of the conductive high-molecular weight material is preferably 0.01 wt%-10.0 wt% based on the entire composition, and especially preferably 0.1 wt%-5.0 wt%. The reason is that if the conductive high-molecular weight material concentration is too low, a large number of discharges will be needed to produce the required film thickness, and mass-production efficiency will be poor, while on the other hand, if the conductive high-molecular weight material concentration is too high, viscosity will be higher.

In preferred practice, the composition for a hole injecting and transporting layer will contain a lubricant. This can effectively prevent drying and solidification of the ink composition in the ink-jet nozzle mouth.

Examples of the lubricant are glycerin, diethylene glycol, and other polyhydric alcohols; glycerin is especially preferred. In preferred practice, the added amount of lubricant is equivalent to about 5 wt%-20 wt% based on the entire composition.

In preferred practice, water, a mixed solvent of water and a lower alcohol, or a mixed solvent of water and a Cellosolve solvent will be used as the polar solvent used for the composition for a hole injecting and transporting layer. The use of these solvents makes it possible to adjust the contact angle of the composition for a hole injecting and transporting layer with respect to the material of the nozzle face of the ink-jet head, the viscosity, and the surface tension without any loss of solubility or dispersibility of the conductive compound. Methanol or ethanol is preferred as the lower alcohol. From the standpoint of film-forming properties, ethoxy ethanol is preferred as the Cellosolve solvent.

Other additives, as well as film stabilizing materials, may be added as well; for example, viscosity modifiers, antioxidants, pH modifiers, preservatives, resin emulsions, leveling agents, etc., may be used.

(Manufacturing process for composition for a hole injecting and transporting layer)

A comparison of film-forming properties and light emission characteristics with and without sonication and a filtration step was conducted using the composition for a hole injecting and transporting layer (Composition 15) given in Table 18, and is given in Table 7. PPV (poly(para-phenylene vinylene)) was used as a green light-emitting layer.

Table 20

Step		Film quality	luminance [cd/m^2]	light emission initial voltage[Vth]	light emission lifetime [hr]
sonication step and filtration step	yes	◎	2000	3.0	5000
	no	Δ	1000	5.0	500

As will be apparent from the preceding results, conducting sonication increases the dispersion, and the use of a composition prepared by filtration of a sonicated dispersion affords a homogeneous hole injecting and transporting layer that has good flatness. In preferred practice, film thickness is 0.05 μm-0.1 μm. The reason is that the film-forming properties of the hole injecting and transporting layer affect the light emission characteristics of the element.

The manufacturing steps for the organic EL element of the present aspect of embodiment are the same as in Aspect of Embodiment 1.

INDUSTRIAL APPLICABILITY

According to the present invention, a composition for a hole injecting and transporting layer is prepared as a liquid, thus permitting optimal design of materials, and the liquid composition is prepared as an ink, thus permitting high-precision patterning of a hole injecting and transporting layer by ink-jet system. Further, the use of a conductive compound, particularly a high-molecular weight material, as the hole injecting and transporting layer material affords manufacture of an organic EL element having high reliability and high qualities.

According to the ink-jet system patterning which pertains to the present invention, there is provided simple, low-cost hole injecting and transporting layer formation.

According to the manufacturing process for an organic EL element which pertains to the present invention, film thickness, number of dots, and other parameters can be adjusted in a desired manner, allowing the size and pattern of a light-emitting element to be

established in a desired manner as well. Further, by combining light-emitting layers of the three primary colors (red, green, blue) produced through ink-jet system patterning, it is possible to develop a highly detailed full-color display with excellent light emission characteristics.

CLAIMS

1 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°.

2 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the viscosity is within the range of 1 cps to 20 cps.

3 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the surface tension is within the range of 20 dyne/cm to 70 dyne/cm.

4 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, and the viscosity is within the range of 1 cps to 20 cps.

5 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the viscosity is within the range of 1 cps to 20 cps, and the surface tension is within the range of 20 dyne/cm to 70 dyne/cm.

6 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the surface tension is within the range of 20 dyne/cm to 70 dyne/cm, and the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°.

7 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, the viscosity is within the range of 1 cps to 20 cps, and the surface tension is within the range of 20 dyne/cm to 70 dyne/cm.

8 The composition for a hole injecting and transporting layer according to any of Claims 1 through 7, wherein the conductive compound is either a high-molecular weight material or a low-molecular weight material.

9 The composition for a hole injecting and transporting layer according to any of Claims 1 through 8, wherein the concentration of the conductive compound is in the range of 0.01 wt% to 10 wt%.

10 The composition for a hole injecting and transporting layer according to any of Claims 1 through 9, wherein the conductive compound is present in a dissolved or dispersed state in a polar solvent as the solvent.

11 The composition for a hole injecting and transporting layer according to Claim 10, wherein the polar solvent is a mixed solvent of water and a lower alcohol.

12 The composition for a hole injecting and transporting layer according to Claim 11, wherein the lower alcohol is methanol or ethanol.

13 The composition for a hole injecting and transporting layer according to Claim 10, wherein the polar solvent is a mixed solvent of water and a Cellosolve solvent.

14 The composition for a hole injecting and transporting layer according to Claim 13, wherein the Cellosolve solvent is ethoxy ethanol.

15 The composition for a hole injecting and transporting layer according to any of Claims 1 through 14, wherein the composition comprises a lubricant.

16 The composition for a hole injecting and transporting layer according to Claim 15, wherein the lubricant is glycerin.

17 A manufacturing process for the composition for a hole injecting and transporting layer according to any of Claims 1 through 16, comprising:

a sonication step; and

a filtration step.

18 A manufacturing process for an organic EL element having a stacked structure of a hole injecting and transporting layer and a light-emitting layer formed

within a partitioning member divided into individual pixel areas, comprising:

a step wherein a partitioning member provided with openings corresponding to pixel areas is formed on a substrate;

a step wherein an ink-jet type recording head is used to fill the aforementioned openings with the composition for a hole injecting and transporting layer according to any of Claims 1 through 17; and

a step wherein the composition for a hole injecting and transporting layer that has filled the openings is subjected to a drying process to form a hole injecting and transporting layer.

19 An organic EL element manufactured by the manufacturing process according to Claim 18.

20 An organic EL element manufactured by the manufacturing process according to Claim 18, wherein the film thickness of the hole injecting and transporting layer is 0.1 μm or less.

21 An organic EL element manufactured by the manufacturing process according to Claim 18, wherein the film resistance of the hole injecting and transporting layer is in the range $0.5 \times 10^9 \Omega/m^2$ to $5 \times 10^9 \Omega/m^2$.

AMENDED CLAIMS

1 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

2 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the viscosity is within the range of 1 cps to 20 cps, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

3 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting

and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the surface tension is within the range of 20 dyne/cm to 70 dyne/cm, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

4 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, the viscosity is within the range of 1 cps to 20 cps, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

5 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the viscosity is within the range of 1 cps to 20 cps, the surface tension is within the range of 20 dyne/cm to 70 dyne/cm, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

6 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the surface tension is within the range of 20 dyne/cm to 70 dyne/cm, the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

7 A composition for a hole injecting and transporting layer for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, comprising:

a conductive compound; and

a solvent;

wherein the viscosity is within the range of 1 cps to 20 cps, the surface tension is within the range of 20 dyne/cm to 70 dyne/cm, the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, and the content of the conductive compound is in the range of 0.01 wt% to 10 wt%.

8 The composition for a hole injecting and transporting layer according to any of Claims 1 through 7, wherein the conductive compound is either a high-molecular weight material or a low-molecular weight material.

9 The composition for a hole injecting and transporting layer according to any of Claims 1 through 8, wherein the conductive compound is present in a dissolved or dispersed state in the solvent as a polar solvent.

10 The composition for a hole injecting and transporting layer according to Claim 9, wherein the polar solvent is a mixed solvent of water and a lower alcohol.

11 The composition for a hole injecting and transporting layer according to Claim 10, wherein the lower alcohol is methanol or ethanol.

12 The composition for a hole injecting and transporting layer according to Claim 9, wherein the polar solvent is a mixed solvent of water and a Cellosolve solvent.

13 The composition for a hole injecting and transporting layer according to Claim 12, wherein the Cellosolve solvent is ethoxy ethanol.

14 The composition for a hole injecting and transporting layer according to any of Claims 1 through 13, further comprising a lubricant.

15 The composition for a hole injecting and transporting layer according to Claim 14, wherein the lubricant is glycerin.

16 A manufacturing process for the composition for a hole injecting and transporting layer according to any of Claims 1 through 15, comprising:

a sonication step; and

a filtration step.

17 A manufacturing process for an organic EL element having a stacked structure of a hole injecting and transporting layer and a light-emitting layer formed within a partitioning member divided into individual pixel areas, comprising:

a step wherein a partitioning member provided with openings corresponding to pixel areas is formed on a substrate;

a step wherein an ink-jet type recording head is used to fill the aforementioned openings with the composition for a hole injecting and transporting layer according to any of Claims 1 through 16; and

a step wherein the composition for a hole injecting and transporting layer that has filled the openings is subjected to a drying process to form a hole injecting and transporting layer.

18 An organic EL element manufactured by the manufacturing process according to Claim 17.

19 An organic EL element manufactured by the manufacturing process according to Claim 17, wherein the film thickness of the hole injecting and transporting layer is 0.1 μm or less.

20 An organic EL element manufactured by the manufacturing process according to Claim 17, wherein the film resistance of the hole injecting and transporting layer is in the range 0.5×10^9 Ω/m^2 to 5×10^9 Ω/m^2.

ABSTRACT

The composition for a hole injecting and transporting layer which pertains to the present invention is a composition for use in patterning formation of a hole injecting and transporting layer of an organic EL element using an ink-jet type recording head, and comprises a conductive compound (the hole injecting and transporting layer component) and a solvent; the contact angle with respect to the material making up the ink discharge nozzle face of the ink-jet type recording head is within the range of 30° to 170°, the viscosity is within the range of 1 cps to 20 cps, and the surface tension is within the range of 20 dyne/cm to 70 dyne/cm. Through these physical characteristics, there is provided a composition for a hole injecting and transporting layer that allows optimization of material and element design to be carried out, and that allows high-precision patterning film formation to be carried out easily, in a short time, and at low cost.

Fig.1A

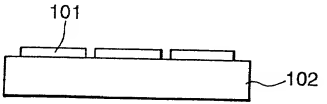


Fig.1B

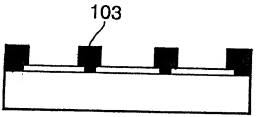


Fig.1C

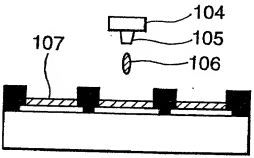


Fig.1D

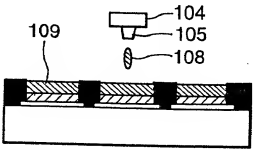
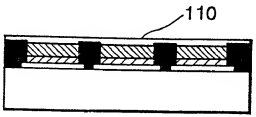


Fig.1E



Seiko Epson Ref. No.: F004113US00 Attorney's Ref. No.: 005317-20009

Declaration and Power of Attorney For Patent Application

特許出願宣言書及び委任状

Japanese Language Declaration

日本語宣言書

下記の氏名の発明者として、私は以下の通り宣言します。

As a below named inventor, I hereby declare that:

私の住所、私書箱、国籍は、下記の私の氏名の後に記載された通りです。

My residence, post office address and citizenship are as stated next to my name.

下記の名称の発明に関して請求範囲に記載され、特許出願している発明内容について、私が最初かつ唯一の発明者（下記の氏名が一つの場合）もしくは最初かつ共同発明者であると（下記の名称が複数の場合）信じています。

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

有機EL素子の製造方法及び有機EL素子

ORGANIC EL ELEMENT MANUFACTURING PROCESS AND ORGANIC EL

上記発明の明細書（下記の欄で×印がついていない場合は、本書に添付）は、

the specification of which is attached hereto unless the following box is checked:

☒ 1999年4月30日に提出され、米国出願番号または特許協定条約 国際出願番号を09/279,483 とし、（該当する場合） _____ に訂正されました。

☒ was filed on April 30, 1999 as United States Application Number or PCT International Application Number 09/279,483 and was amended on _____ (if applicable).

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I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

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私は、米国法典第３５編１１９条（ａ）-（ｄ）項又は３６５条（ｂ）項に基き下記の、米国以外の国の少なくとも１ヶ国を指定している特許協力条約３６５条(ａ)項に基づく国際出願、又は外国での特許出願もしくは発明者証の出願についての外国優先権をここに主張するとともに、優先権を主張している、本出願の前に出願された特許または発明者証の外国出願を以下に、枠内をマークすることで、示しています。

I hereby claim foreign priority under Title 35, United States Code, Section 119 (a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s) 外国での先行出願			Priority Not Claimed 優先権主張なし
9-237103	Japan	02/September/1997	☐
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	
10-214596	Japan	29/July/1998	☐
(Number) (番号)	(Country) (国名)	(Day/Month/Year Filed) (出願年月日)	

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I hereby claim the benefit under Title 35, United States Code, Section 119 (e) of any United States provisional application(s) listed below.

(Application No.) (出願番号)	(Filing Date) (出願日)	(Application No.) (出願番号)	(Filing Date) (出願日)

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PCT/JP98/03935	02/September/1998	Pending
(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況：特許許可済、係属中、放棄済)
(Application No.) (出願番号)	(Filing Date) (出願日)	(Status: Patented, Pending, Abandoned) (現況：特許許可済、係属中、放棄済)

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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William Wright (Reg. 36,312)
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John P. Scherlacher (Reg. 23,009)
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15

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

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